QUALITY ASSURANCE FOR MEASUREMENTS OF THE RADIOACTIVITY IN THE AREA OF THE"HORIA HULUBEI" NATIONAL INSTITUTE FOR PHYSICS AND NUCLEAR ENGINEERING, IFIN- HH

Ana Stochioiu, Aurelian Luca, Maria Sahagia, Romul Mircea Margineanu, Ion Tudor

"Horia Hulubei" National Institute for R&D in Physics and Nuclear Engineering, IFIN-HH
Bucharest, Magurele, Romania

Abstract – This paper presents one part of the activities deployed by the Laboratory for Environment and Personnel Dosimetry (LDPM) of IFIN-HH, namely the radiological monitoring of the environment within the Institute's area and its surrounding influence zone, according to the program approved by the national regulatory body for nuclear activities, CNCAN. The representative reports regard the radioactive content of soil, surface and underground water, cultivated and spontaneous vegetation, aerosols and atmospheric fallout, sediments. The common requirement is that the measured quantities be precise and the reported values be reliable and credible. This goal is achieved by maintaining a Quality System, verified within the obtaining and maintaining of the laboratory accreditation, according the international standard ISO/IEC 17025:2005.The LDPM is accredited by the Romanian accreditation body, RENAR, member of the European Accreditation, EA and is designed by CNCAN as a notified testing laboratory. Many measurements were performed in collaboration with the Radionuclide Metrology Laboratory (RML) from IFIN-HH, RENAR accredited and CNCAN notified for calibration and for testing in the field of radioactivity measurement. This paper proposes a short presentation of the important aspects in our activity:
(i) description of equipment, sampling methods, processing and measurement of environmental samples;
(ii) validation of equipment and methods by participation in international and national proficiency tests;
(iii) a five year follow chart, containing the results in measurement of samples;
(iv) a recent application, with a wide impact in Romanian mass media: the credible daily report on the possible influence of Fukushima accident over the Romanian environmental radioactivity

Keywords: monitoring; environment, radioactivity

1. EQUIPMENT AND METHODS

The main specific quantities describing an environment sample are: a) Specific activity of solid samples (Am = A/m) and b) Specific activity of liquids and air (Av = A/v), where: Am is the specific mass activity, Av is the specific volume activity, m is the solid sample mass, v is the liquid or air samples' volumes, A is the total activity of the sample [1].
The equipments notified for these measuring methods are:

- the gamma-ray spectrometer, installed at RML, containing a Hyper-Pure Germanium semiconductor detector (HPGe) ORTEC (USA), Model GEM25P4, a specially designed shield, consisting from: 10 cm lead, 1 mm cadmium and 2 mm copper, and the spectral analysis system; operation and analysis software, like: coincidence summing corrections and efficiency transfer coefficients for various geometries and densities (GESPECOR) [2], background subtraction; GammaVision-32, the manufacturer software used for spectra acquisition) [3], deconvolution of multiple peaks (COLEGRAM) [4] and decay corrections from the measurement to the PT/ILC reference date.
- the gamma –ray spectrometer with high resolution GEM 25 P4 HPGe detector
- the 9300-PG, GFR, PROTEAN equipment for the gross alpha, beta activity measurement;
- the 55-XLB-6 equipment, which automatic sample changer, for the gross alpha, gamma, activity measurement
- the aerosol sampling pump with a F&Y model 521147 filter.
- analytical balance Model WAX 220, PARTNER;
- equipment for primary processing of samples: evaporation, drying and calcinations.

The principal method used for the sampling is described in detail in the laboratory work procedures, which is part of the Quality Management System (QMS) of the LDPM.
The methods for their measurement are basically grouped in two categories:
I. The gross (alpha, beta)/(and alpha, beta, gamma) activity measurement.
II. The gamma-ray spectrometry analysis.
The metrological traceability of the whole equipment to a primary standard laboratory is assured by using radioactive standards provided by the IFIN-HH, Radionuclide Metrology Laboratory (RML), primary laboratory, equivalent at the CIPM-MRA level via the KCDB records and BIPM – JCRB approved CMC files.

2. VALIDATION OF THE EQUIPMENT AND METHODS

In order to validate the results, the laboratory participated at a series of international and national proficiency tests, organized by prestigious laboratories.

2.1 Environmental radioactivity proficiency test exercise 2008, organized by the National Physical Laboratory (NPL), UK.

The samples were prepared by the NPL and consisted in mixed-radionuclide solutions (sources prepared from single radionuclide standard solutions, traceable to NPL, UK primary standard) – GH samples and neutron activated crushed concrete (solid powder) – C samples, AH (high level of activity) and AL (low level of activity) samples (liquid sample) for gross alpha and B sample for gross beta determination.

Thin samples were prepared in standard supports with the diameter of 50 mm. The measurements in the domain of gross alpha (a) and beta (b) were done with 9300-PG, GFR, PROTEAN equipment ; the detection limits were: 0.010 Bq for gross alfa measurements and 0.029 Bq for gross beta measurements.

The results, with the corresponding uncertainty values [66(3) means 66±3)] and the comparison to the reference values provided by the organizers were published in the NPL Report IR 15/2009 [5] and are presented in the Table 1:

TABLE 1.
Gross alpha and beta measurement results

Sample	IFIN-HH Result	NPL - Assigned Result	IFIN-HH Deviation (%)	z-score
Gross a AL , Bq kg-1	66(3)	92(8)	-28(7)	-5.49
Gross a AH , Bq g-1	53.0(21)	65(8)	-19(11)	-3.76
Gross b B2, Bq g-1	1.87(9)	1.1423(23)	64(8)	10.64
Gross b Ccrushed, Bq g-1	1.23(6)	1.2(8)	1(8)·101	0.10

It can be seen that except the gross b B2 sample, the results are acceptable from the point of view of the z-score.

The second part of the measurement regarded the gamma-ray analysis of the GH and C type samples, performed in the RML, by the gamma-ray spectrometry method; the corresponding uncertainty values and the comparison with the reference values given by the organizers (NPL), are presented in the Table 2, reference [5]:

TABLE 2.
Individual radionuclide concentrations of samples

Radionuclide, sample type	IFIN-HH Result (Bq g-1)	NPL Assigned Result (Bq g-1)	IFIN-HH Deviation (%)	z-score
^{22}Na GH	4.73(13)	5.529(20)	-14.5(24)	-3.58
^{60}Co GH	3.90(11)	4.641(14)	-16.0(24)	-4.41
^{95}Zr GH	6.16(16)	7.35(8)	-16.2(23)	-4.13
^{95}Nb GH	11.3(5)	13.54(7)	-17(4)	-3.39
^{133}Ba GH	1.98(7)	2.754(19)	-28(3)	-6.22
^{134}Cs GH	3.6(4)	4.63(4)	-21(7)	-5.46
^{137}Cs GH	7.90(22)	9.56(7)	-17.4(24)	-4.59
^{152}Eu GH	14.3(4)	17.86(12)	-20.0(23)	-5.15
^{60}Co C	0.114(6)	0.1045 (14)	9(6)	1.73
^{133}Ba C	0.0059 (19)	0.0070(3)	-20(30)	-1.30
^{152}Eu C	2.9(1)	2.63(4)	10(4)	2.36
^{154}Eu C	0.114(1)	0.1029 (18)	11(10)	1.40

In this case many results were reported and they are in almost cases acceptable from the point of view of z-score.

2.2 The JRC-IRMM, CCRI(II)-S8 Supplementary Comparison on 40K, 137Cs and 90Sr activity content in dried bilberry material (2010-2011)

Two series of six bilberry samples (about 100 g each) were prepared and distributed by the JRC- Institute for Reference Materials and Measurements (IRMM), Geel - Belgium. They were measured at IFIN-HH, RML, with the purpose of characterizing a reference material within a supplementary comparison type CCRI(II)-S8, and at LDPM. The activity concentrations (Bq/kg dry mass) were reported; a special drying procedure, according to the instructions of the organizers, was applied, in order to determine the water content for each sample.

The measurements performed at RML regarded the determination of ^{40}K and ^{137}Cs content.
Six bilberry samples (about 100 g each) were measured, and the activity concentrations (Bq/kg dry mass) of ^{137}Cs and ^{40}K, determined by high resolution gamma-ray spectrometry, were reported; the reference date was 01/01/2009. The relative water content of the six samples, determined by using small sample aliquots (each with a mass of about 0.5 g) was between 3.8% and 4.3%.
The results of all the participants are under evaluation by the organizers; the preliminary results will be discussed and presented in a paper accepted at the ICRM2011 International Conference, in September 2011 [6].
The measurements done at LDPM regarded the determination of 90Sr content in the mixture. As it is well known, ^{90}Sr is a pure beta decaying radionuclide; a secular equilibrium is established with its daughter, ^{90}Y, a pure beta emitter, too. Its content is measured by a standard method consisting from the chemical separation from the sample and the measurement of the pure ^{90}(Sr+Y) sample in equilibrium conditions. The laboratory does not dispose of a radiochemistry laboratory for chemical separation and an alternative, indirect method was used. It consisted from the followings. A series of 12 samples of 0.500 g were gravimetrically prepared and then dried according to the procedure indicated by the organizer. The gross beta activity was measured for each sample, using the 9300-PG, GFR, PROTEAN equipment for the gross alpha, beta activity measurement, calibrated with a ^{90}(Sr+Y) standard source; the detection efficiency was (0.425±0.023) impulses/(Bq s). The contributions in beta counting rate of ^{40}K and ^{137}Cs, beta-gamma ray emitters, were calculated from the activity concentration measurement by gamma-ray spectrometry and the decay scheme parameters; their detection efficiency was approximated as equal to that of ^{90}(Sr+Y). Due to the high content of the two radionuclides from the mixture, ^{40}K and ^{137}Cs, the uncertainties were high, about 20%. The result could not be taken into account for the Supplementary comparison. However, the determination of ^{90}Sr was a useful exercise for testing the normal functioning of the equipment and the professional skills of the laboratory staff; it was also an opportunity to explore the limits of the indirect activity determination method.

2.3 Proficiency test organized by IFIN-HH, RML

Both gamma-ray spectrometers were tested through a proficiency test organized by the RML with the accredited/notified IFIN-HH laboratories performing gamma-ray spectrometry measurements.
It consisted from the measurement of a volume sample, water equivalent, containing a mixture of ^{134}Cs + ^{137}Cs. The reference value was certified by the IFIN-HH, RML. The results are analysed in the papers [7, 8]. As a general conclusion, while both spectrometer collectives obtained satisfactory results in the measurement of ^{137}Cs, the result of the RML measurement of ^{134}Cs is better than in LDPM spectrometer case, due to a more appropriate choice of the standard source used for efficiency calibration. The deviations of RML (testing laboratory) and LDPM ^{134}Cs activity results from the reference value are -0.79 %, respectively -15 %.

3. A FIVE YEAR FOLLOW UP OF THE CONTENT OF RADIONUCLIDES IN SOIL, VEGETATION AND MILK AROUND IFIN-HH AREA

The gamma-ray spectrometry analysis, using the LDPM spectrometer, was done for the samples. The mean annual values of the specific and volume activities of ^{137}Cs and ^{40}K, expressed in Bq kg-1 (Bq l-1 for milk), during the period 2008-2010, for soil, vegetation and milk samples, measured by gamma-ray spectrometry are presented in the Table 3, such as they were annually reported [9]. The reported uncertainties are standard deviations of the mean, calculated from 9 points and monthly sampling per year.

TABLE 3. Mean annual concentrations of ^{137}Cs and ^{40}K

Sample/ Year	2008		2009		2010	
	^{137}Cs	^{40}K	^{137}Cs	^{40}K	^{137}Cs	^{40}K
Soil	55.9 ±5.9	640 ±34	40.5 ±3.8	638 ±29	44.4 ±3.3	585 ±17
Spont Veget.	7.2 ±1.2	21.7 ±3.7	6.0 ±1.8	25.5 ±4.7	6.3 ±2.1	22.6 ±0.2
Cultiv. Veget.	3.6 ±0.7	22.7 ±2.9	5.8 ±1.7	20.0 ±1.1	4.2 ±1.3	24.2 ±4.3
Milk	1.8 ±0.1	32.6 ±4.1	1.3 ±0.2	29.0 ±3.2	1.1 ±0.1	36.0 ±2.9

The following conclusions can be drawn from the table 3:
- A common comment is that the only identified radionuclides, included in this table, are ^{40}K, natural radionuclide and ^{137}Cs still existing after 25 years since the Chernobyl nuclear accident. No artificial radionuclides resulting from IFIN-HH nuclear activities were detected.
- The content of the natural ^{40}K remains constant, within the limits of annual variation, while ^{137}Cs presents a slight decrease.

4. MONITORING OF THE FUKUSHIMA ACCIDENT IMPACT OVER THE IFIN-HH AREA

The air monitoring during the year 2011 and the evaluation of the Fukushima accident impact
The method refers at the quantitative aspiration of a 20 m^3 of air; the aerosols are retained on the quantitative filter, yield (96-98)%. The gross alpha and beta activity is measured in three steps: 3 min. after aspiration, to detect the ^{222}Rn daughters content, 20 h for the

measurement of ^{220}Rn chain and 5 days after the aspiration time, to detect artificial, long lived radionuclides [10].

The mean volume activity in 2010 was: (128 ± 14) mBq m-3 after 20 h from prelevement, due to 220Rn daughters, and (5.00 ± 2.00) mBq m-3 after 5 days, due to long lived artificial radionuclides. For comparison purposes, the mean values recorded during the year 2011, comparatively, for January, February and 4 weeks in March are presented in Table 4.

TABLE 4. The mean values recorded during the year 2011

Month	January	February	March, 1-8 week
Mean value, mBq m^{-3}, 20 h	145±11	155±14	115.7±6.7
Mean value, mBq m^{-3}, 5 d	5.9±2.0	6.7±1.2	5.5±1.7
Month	March, 9-16 week	March, 17-24 week	March, 25-31 week
Mean value, mBq m^{-3}, 20 h	114.3±4.2	89.2±6.1	121.2±6.1
Mean value, mBq m^{-3}, 5 d	5.9±1.3	6.3±1.1	5.0±1.2

From the above presented results, one may conclude that the values measured at IFIN-HH, during March 2011, are situated within the limits of those registered during the year 2010 and in January – February 2011. No influence of the Fukushima accident can be detected. These data were offered to the public via the site www.nipne.ro and to the Romanian mass media, under the title of official IFIN-HH dosimetry bulletins.

CONCLUSIONS

The LDPM, IFIN-HH, accomplishes his duties of following the radioactivity of environment and food chain samples and reporting its levels, according to the legislation.

The equipment and methods are adequate for these purposes, being traceable to a primary standard.

The technical capability was proved by the participation, with satisfactory results, at international and national proficiency tests.

The multiannual follow of some samples showed the existence of only natural ^{40}K and Chernobyl origin ^{137}Cs, no influence of IFIN-HH activities being detected.

The Fukushima nuclear accident had no radiological impact over the IFIN-HH area.

REFERENCES

[1] Guidelines for Calculating Derived Release Limits for Radionuclides in Airborne and Liquid effluents for Whiteshell nuclear establishment, AECL 10028, 1989;

[2] GESPECOR version 4.2. Manual, CID Media GmbH 2007, Germany

[3] ORTEC GammaVision-32, version 6.01: HPGe Gamma Ray Spectrum Analysis and MCA Emulation. User Manual, AMETEK, Inc., June 2007, USA.

[4] H. Ruellan, M.C. Lepy and J. Plagnard. Presentation du logiciel "COLEGRAM", Note Technique LPRI/95/016, 1995, Saclay, France.

[5] A. Harms and C. Gilligan, NPL Report IR 15: Environ mental radioactivity proficiency test exercise 2008, May 2009, NPL, Teddington, UK.

[6] U. Wätjen, Y. Spasova, Z. Szántó, H. Emteborg, T. Altzitzoglou, P. Oropesa, L. García, Y. Moreno, A. Ceccatelli, A. Luca, C. Frechou, L. Ferreux , S. Pierre, L. Szücs, J. La Rosa, H. Wershofen, M. Schmiedel, K. Trothe, Ü. Yücel, H. Dikmen, Y.Ö. Özkök. Results of an international comparison for the determination of radionuclide activity in bilberry material. 18-th International Conference on Radionuclide Metrology and its Applications (ICRM2011), Tsukuba, Japan, 19 -23 September 2011.

[7] M. Sahagia, A. Antohe, A. Luca. Results of a proficiency test on the measurement of volume radioactive sourcea from the mixture: ^{134}Cs ^{137}Cs. 3-rd International Conference on Proficiency Testing, Iasi, Romania, 28-30 September 2011

[8] A. Luca, M. Sahagia, A. Antohe, C. Şulea, R.Berenştain. Recent participations of the Radionuclide Metrology Laboratory from IFIN-HH to profiviency tests and inter-laboratory comparisons. 3-rd International Conference on Proficiency Testing, Iasi, Romania, 28-30 September 2011.

[9] A. Stochioiu - Reports on the Environmental Radioactivity for IFIN-HH: 2006, 2007, 2008, 2009 and 2010.

[10] A. Stochioiu, M. Sahagia, S. Bercea, C. Ivan, I. Tudor. Monitoring of the radioactivity concentration of air in the area of IFIN-HH, Romania. Rom. J. Physics 61, 3 (2009) 581-586.

Ana Stochioiu - "Horia Hulubei" National Institute for R&D in Physics and Nuclear Engineering, IFIN-HH, The Department of Life and Environmental Sciencies, 30 Reactorului str, Magurele Bucharest, , Romania, e-mail: stoc@nipne.ro, tel: 0040214046224; fax: 0040214046190

Aurelian Luca - "Horia Hulubei" National Institute for R&D in Physics and Nuclear Engineering, IFIN-HH, , Radioisotopes and Radiation Metrology Department, 30 Reactorului str, Magurele Bucharest, , Romania, e-mail: aluca@nipne.ro tel: 0040214046224; fax: 0040214046190

Maria Sahagia - "Horia Hulubei" National Institute for R&D in Physics and Nuclear Engineering, IFIN-HH, Radioisotopes and Radiation Metrology Department, 30 Reactorului str, Magurele Bucharest, , Romania, e-mail: msahagia@nipne.ro , tel: 0040214046224; fax: 0040214046190

Romul Mircea Margineanu - "Horia Hulubei" National Institute for R&D in Physics and Nuclear Engineering, IFIN-HH, -HH, The Department of Life and Environmental Sciencies, 30 Reactorului str, Magurele Bucharest, , Romania, tel: 0040214046224; fax: 0040214046190

Ion Tudor - "Horia Hulubei" National Institute for R&D in Physics and Nuclear Engineering, IFIN-HH, -HH, The Department of Life and Environmental Sciencies, 30 Reactorului str, Magurele Bucharest, , Romania, e-mail: itudor@nipne.ro, tel: 0040214046224; fax: 0040214046190